Ceramic-Metal Interfaces in Nuclear Materials Applications[1]

by

S. M. McDeavitt

Argonne National Laboratory
Chemical Technology Division
9700 S. Cass Ave.
Argonne, IL 60439

Phone: (630) 252-4308
Fax: (630) 252-9917
e-mail: mcdeavitt@cmt.anl.gov

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Argonne National Laboratory
Chemical Technology Division
Argonne, IL 60439-4837

Nuclear fuels and fuel processing technologies are typically beset with significant materials performance challenges due to vigorous chemical environments and elevated temperatures. In addition, many of the Generation IV (GenIV) nuclear systems on the horizon push the performance envelope by assuming marked improvements in high temperature materials. Therefore, the high-temperature durability of materials in aggressive environments is of paramount importance to advanced nuclear systems and processing methods. Advanced ceramics, coatings, and functional graded materials are all likely to find utility in these advanced nuclear technologies. The arena of ceramic-metal interfacial chemistry therefore becomes a critical issue for the success of some GenIV concepts.

To explore this idea, a database of high-temperature molten metal-stable ceramic interaction studies is being reviewed [1-3]. This data was generated during the development of advanced crucible materials for processing uranium metal and stainless steel-zirconium waste form alloys for the electrometallurgical treatment of spent nuclear fuel from the Experimental Breeder Reactor-II. A series of sessile-drop style experiments was conducted to evaluate the ceramic-metal interface between molten uranium and zirconium alloys and candidate ceramics. The issues surrounding the observed interface reactions and the materials engineering solutions designed to minimize them are in the same fundamental materials science category as ceramic joining, even though the applications are fundamentally different.

Figure 1 shows a listing of the initial ceramic materials screened by this method and a diagram of the experimental setup. In general, the interaction between the liquid metal drop and the ceramic substrate may be broken into three categories: (1) complete non-wetting, (2) reactive wetting, and (3) vigorous chemical interaction. The goal of the screening exercise was to identify materials in the complete non-wetting category; a number of hafnium-based ceramics were selected and the program moved on toward the development of fabrication methods for crucibles based the HfN and $Hf_2Y_2O_7$ compounds.

In the context of GenIV materials, these reactive wetting results may be relevant to the design of reactor material surfaces and the development of reactive filler metals for the fabrication of high temperature reactor materials through metal-ceramic joining. Reactive wetting involves the chemical alteration of the metal-ceramic interface to enable liquid spreading. Parameters that have marked impact on this interface reaction include the thermodynamic stability of the substrate, the properties of the modified interface, the temperature-dependent solubility limits of the liquid and solid phases, and the high-temperature stoichiometry of the ceramic. In the noted database, four general classifications of the interface features may be made:

(1) No observable reaction product phase between the liquid metal and the ceramic with minimal chemical exchange between the phases,

(2) Observable reaction product phase between the liquid metal and the ceramic with minimal chemical exchange between the phases,

(3) Observable reaction product phase between the liquid metal and the ceramic with extensive chemical exchange between the phases, and

(4) No observable reaction product phase between the liquid metal and the ceramic with extensive chemical exchange between the phases (by dissolution and infiltration).

Figure 2 shows an example of the third category where molten zirconium was in contact with zirconium nitride. In this particular case, the liquid drop of zirconium reduced the ZrN substrate and became saturated with nitrogen. A layer of substoichiometric ZrN (or ZrN_{1-x}) was formed at the interface and additional ZrN_{1-x} phases formed on cooling and are observed at the grain boundaries in the zirconium drop. This particular example highlights the role of temperature-dependent solubility limits of the liquid on reactive wetting.

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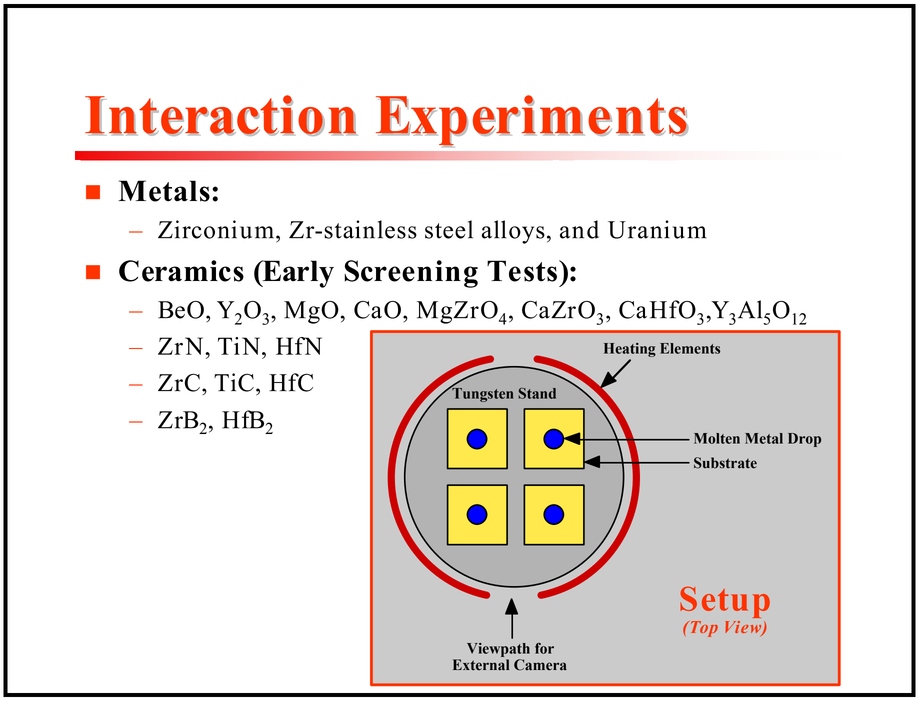

Figure 1. Experiment Conditions and Initial Materials Selection for High Temperature Interaction Studies.

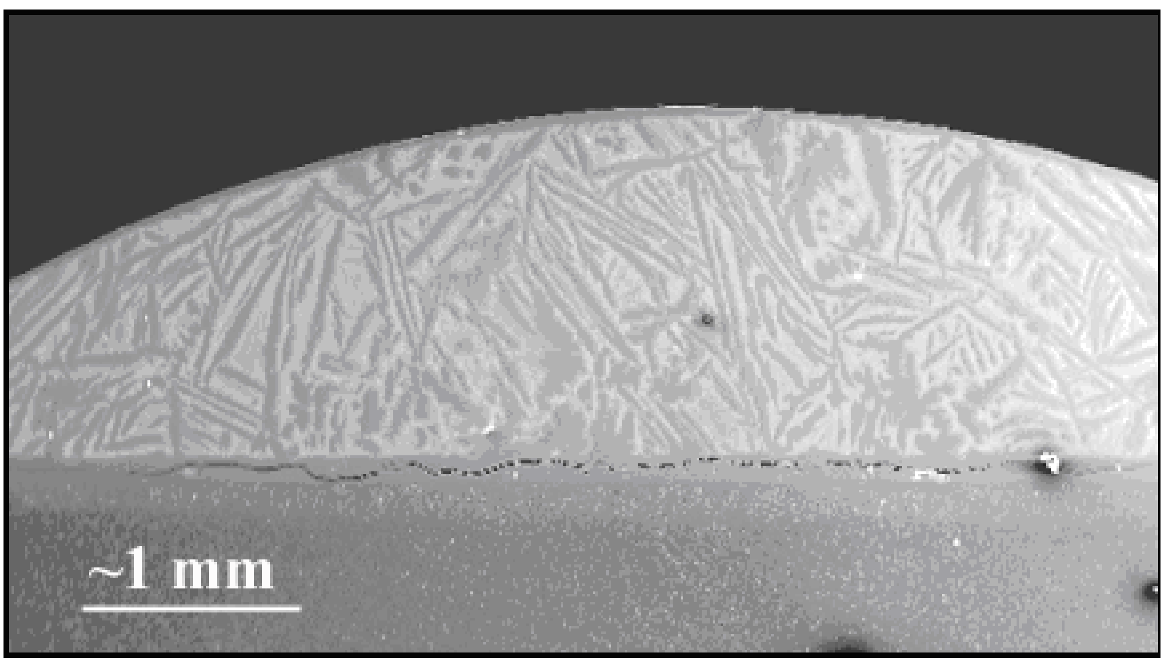

Figure 2. Interface Developed Between Molten Zr and ZrN at ~2000°C (visible ZrN_{1-x} formed on cooling).